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Uvarov, I. V.; Postnikov, A. V.; Shlepakov, P. S.; Naumov, V. V.; Koroleva, O. M.; Izyumov, M. O.; Svetovoy, V. B.

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Fast electrochemical membrane actuator: Design, fabrication and preliminary testing

I V Uvarov[1], A V Postnikov[1], P S Shlepakov[1], V V Naumov[1], O M Koroleva[1], M O Izyumov[1] and V B Svetovoy[1,2]

[1]Yaroslavl Branch of the Institute of Physics and Technology RAS, 150007, Universitetskaya Street 21, Yaroslavl, Russia
[2]Zernike Institute for Advanced Materials, University of Groningen - Nijenborgh 4, 9747 AG Groningen, The Netherlands

E-mail: i.v.uvarov@bk.ru

Abstract. An actuator based on water electrolysis with a fast change of voltage polarity is presented. It demonstrates a new actuation principle allowing significant increase the operation frequency of the device due to fast termination of the produced gas. The actuator consists of a working chamber with metallic electrodes and supplying channels filled with an electrolyte. The chamber is formed in a layer of SU-8 and covered by a flexible polydimethylsiloxane membrane, which deforms as the pressure in the chamber increases. Design, fabrication procedure, and first tests of the actuator are described.

1. Introduction

Membrane actuator is a wide spread component of microfluidic systems. It is a key part of pumps [1], valves [2], lenses, mirrors [3] and other devices. Several mechanisms driving the membrane motion are known: pneumatic, piezoelectric, electromagnetic and others [4]. Typically the actuator is integrated with other microfluidic components, so its structure should be as simple as possible. Electrochemical actuation is very attractive because it allows a simple design and provides a strong response. It is based on the electrochemical gas production with the following termination of the gas. The main disadvantage of the electrochemical actuators is a long response time due to slow recombination of the gas inside of the working chamber [1]. Recently a short-time electrolysis using alternating polarity voltage pulses was demonstrated, which allows fast generation and termination of the gas in the chamber [5]. Here we describe design, fabrication, and first results of testing of the electrochemical membrane actuator working on the new actuation principle.

2. Design and operation principle of the actuator

Design of the actuator is schematically shown in figure 1. Actuator has a chamber with metallic electrodes inside of it, channels for filling the system with the electrolyte, and holes for connection with microfluidic tubes. The diameter of the chamber is 500 μm and the height is 8 μm. An oxidized silicon substrate is used as the bottom wall, the side walls are made of cured SU-8 resist, and the upper wall is the flexible polydimethylsiloxane (PDMS) membrane with a thickness of 30 μm. The actuator has a planar configuration, the chamber and the channels have the same height. The electrolyte is a molar solution of Na_2SO_4 in distilled water. Photograph of the fabricated actuator with close-up view of the working chamber is shown in figure 2. The electrodes have interdigitated structure to increase

the efficiency of the electrolytic bubble generation [6]. The diameter of the overall electrode area is 450 μm, which is close to the diameter of the working chamber. Interdigitated structure of electrodes and their size are chosen to saturate the entire volume of the chamber evenly with the gas bubbles.

Operation principle of the actuator is schematically shown in figure 3. A series of short (~10 μs) voltage pulses of alternating polarity is applied to the working electrode (the other one is grounded). These pulses produce hydrogen and oxygen due to water electrolysis. The alternating polarity regime differs in that sense that all the gas is collected in nanobubbles, which are invisible because they do not scatter light [7]. The pressure increases in the chamber and pushes the membrane up. When the pulses are switched off, the gas in nanobubbles recombines into water in 1 ms or so releasing the pressure and the membrane returns to its initial state. Unlike the conventional operation mode when a single polarity voltage pulse is applied to the electrodes, in the alternating polarity mode hydrogen and oxygen molecules are not spatially separated and form not only H_2 and O_2 bubbles, but also bubbles containing mixtures of H_2 and O_2 [5]. The latter disappear in the reaction, which is ignited in nanobubbles spontaneously. The mechanism of the reaction is still not completely clear but the reaction happens with the assistance of the gas-liquid interface [5]. When the pulses are switched off the pressure is reduced in about 1 ms. Short recombination time makes it possible to reduce significantly the duration of the actuation cycle.

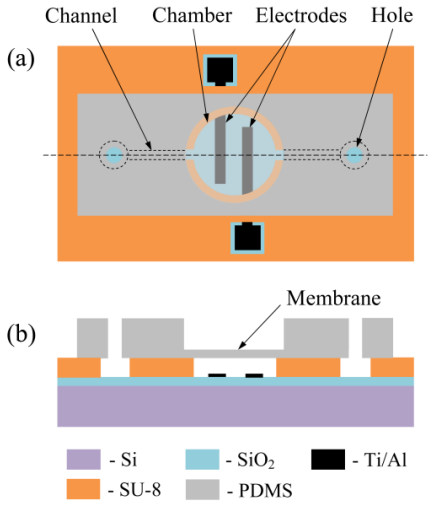

Figure 1. Design of the actuator: top view (a) and the cross-section (b).

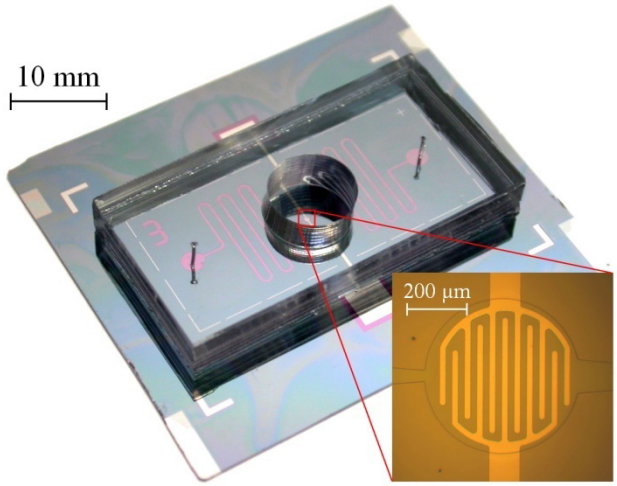

Figure 2. Ready to use actuator. The hole in PDMS layer is much larger than the working chamber covered with the membrane. The inset shows a zoomed view of the chamber.

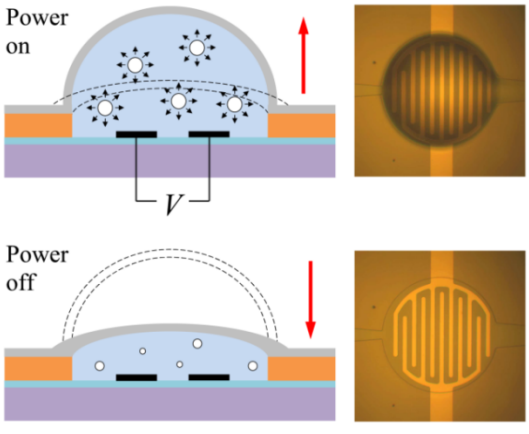

Figure 3. Operation principle of the actuator. Gas produced in the chamber electrochemically pushes the membrane up. When the voltage is switched off the gas in nanobubbles is terminated fast and the membrane is going down.

3. Fabrication

The main steps of the fabrication process are shown in figure 4. The process includes three stages. At the first stage a silicon substrate containing metallic electrodes and a layer of SU-8 containing the chamber and channels is prepared. The wafer is thermally oxidized in wet oxygen so that a dielectric layer of a thickness of 0.9 μm is formed on the wafer (figure 4a). Next, the electrodes of the actuator are fabricated (figure 4b). The electrodes contain 10 nm thick adhesive Ti layer, 500 nm thick Al layer, and 100 nm thick Ti layer deposited by magnetron sputtering. The substrate is covered by the photoresist Shipley S1813 and the UV contact photolithography is performed followed by the wet etching of Ti/Al/Ti layers through the photoresistive mask. Further, the walls of the chamber and channels are formed in 8 μm thick layer of the negative photoresist SU-8 3005 (figure 4c). The post-exposure baking of SU-8 is done at a temperature no higher than 95 °C in order to preserve the residual epoxy groups on the SU-8 surface [8].

At the second stage a PDMS structure holding a membrane is fabricated. The silicon wafer is treated in hydrofluoric acid solution for 10 s to make its surface hydrophobic. It is necessary to reduce the adhesion of PDMS to silicon. A PDMS layer with a thickness of 30 μm is applied to the wafer by spin-coating of liquid compound (Sylgard 184, a mixture of a base and a curing agent at 10:1 ratio) and curing at the temperature of 100 °C during 35 min (figure 4d). Then 4 mm thick PDMS structure with a through hole of 8 mm in diameter is formed separately and bonded to the thin PDMS layer (figure 4e). The bonding is performed using oxygen plasma treatment of PDMS at the pressure of 0.5 mbar and 300 W power during 8 s. Next, the PDMS structure is detached from the wafer with the through hole being closed by a membrane. After that the inlet and outlet holes with a diameter of 0.5 mm are made in the structure by a biopsy puncher (figure 4f).

At the final stage the PDMS structure containing the membrane and the silicon substrate containing the electrodes, chamber, and channels are assembled together using irreversible bonding of PDMS to SU-8. The bonding is based on the chemical reaction between PDMS and SU-8 by generating amino groups on PDMS surface with nitrogen plasma treatment and allowing the amino groups to react with the residual epoxy groups on SU-8 surface at elevated temperatures [8]. The PDMS structure is exposed to N_2 plasma with the pressure of 0.5 mbar and 700 W power during 1 s. Exposed surface is brought into contact with the SU-8 surface aligning the structure in such a way that the chamber is covered by the membrane and the channels are sealed with a thick layer of PDMS (figure 4g). The assembled parts are treated at 100 °C for 30 min to drive the chemical reaction. After cooling the samples are ready for filling with the electrolyte and testing.

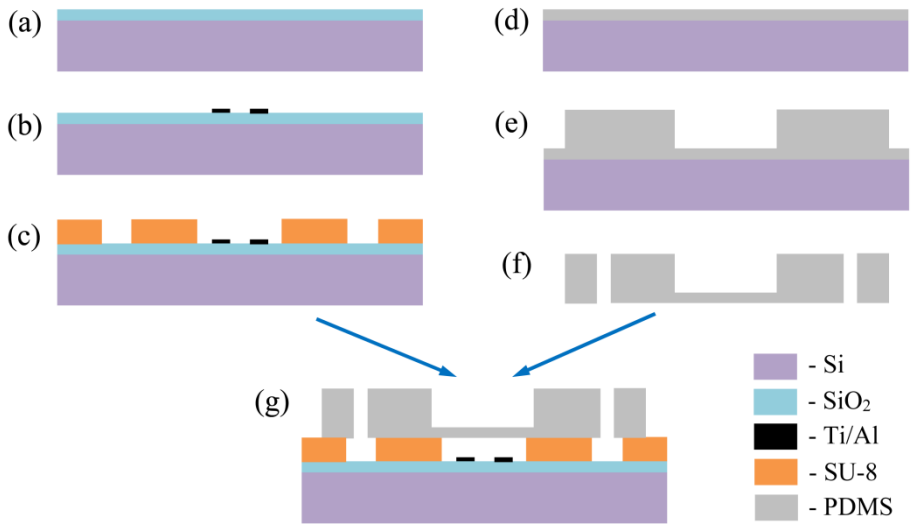

Figure 4. Fabrication procedure.

4. Preliminary testing of the actuator

Performance of the actuator is measured using a homemade Fabry-Perot interferometer. The PDMS membrane has poor reflectivity even after deposition of a semitransparent metallic layer. Laser beam passing through and reflecting from the bottom wall of the chamber creates parasitic interference. To avoid this, a thin rectangular piece of silicon acting as a working mirror is mounted on the membrane in such a way that the membrane deflection tilts the piece as shown in figure 5. The mirror has lateral dimensions of 2×1.5 mm^2, a thickness of 30 μm, and a mass of 0.2 mg. Photograph illustrating the position of the working mirror on the actuator is shown in figure 6. The edge of the mirror is located as close as possible to the center of the membrane to increase the accuracy of the measurement.

The interference pattern is a system of stripes of equal thickness created by interfering laser beams (wavelength $\lambda = 630$ nm) reflected from the reference and working mirrors. The pattern is observed by a microscope. An example of stripes without applying the driving voltage is shown in figure 7a. A series of pulses applied to the electrodes deforms the membrane and changes the angle between the mirrors increasing the number of stripes (figure 7b). During the test the pattern is recorded with a framerate of 25 fps by the video camera ToupCam U3CMOS03100KPA mounted on the eyepiece of the microscope. The deflection of the membrane from the initial state d at each frame can be calculated as follows:

$$d = \frac{L}{l}\frac{\lambda}{2}(N_2 - N_1),$$

where $L = 2$ mm is the length of the working mirror, $l = 0.8$ mm is the part of the mirror captured by the frame, N_1 and N_2 are numbers of stripes at the initial state and at a given frame, respectively.

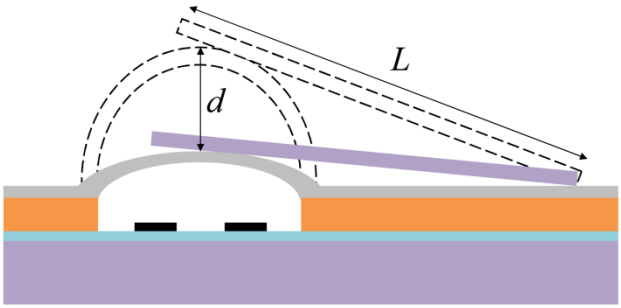

Figure 5. Schematic side view of the working mirror mounted on the actuator. Dashed lines indicate the position of the membrane and the mirror when a series of pulses is applied to the device.

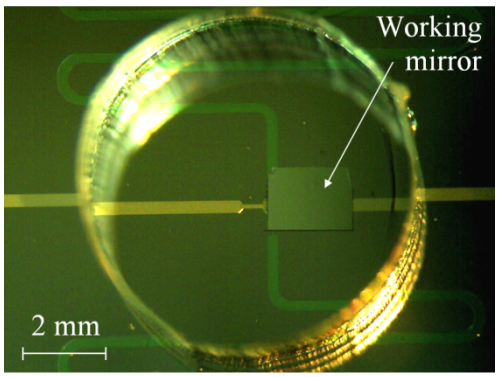

Figure 6. Photograph of the actuator with the working mirror installed (top view).

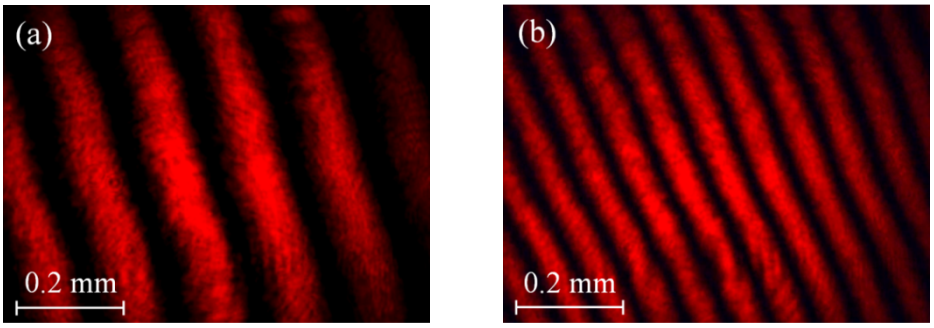

Figure 7. The interference pattern: initial state (a) and the state with the deformed membrane (b).

To drive the actuator a series of $N = 200000$ rectangular voltage pulses of alternating polarity with a duration of $t_0 = 1.25$ μs (frequency of pulses $f = 400$ kHz) is applied to the electrodes. Duration of the series is $t_s = Nt_0 = 250$ ms. Maximal deflection of the membrane as a function of the pulses amplitude U is shown in figure 8. The highest deflection of 7.6 μm is obtained at $U = 13.5$ V. This deflection corresponds to an increase in the chamber volume by approximately 1.5 times in comparison with the nominal value (1.6 nl). Further increase in the amplitude does not lead to larger membrane deflection probably because of the formation of pinned microbubbles in the working chamber. Unfortunately, the working mirror does not allow to observe the chamber. Relatively high amplitude of pulses is needed to drive the actuator that is explained by the formation of TiO_2 on the electrodes [9].

Deflection of the membrane during several actuation cycles at $U = 13.5$ V is shown in figure 9. The time interval between series of pulses is 3-5 s. The membrane reaches the maximal deflection for less than 200 ms and returns to the initial state first very fast but then the process slows down. The total relaxation time is approximately 5 s so that the actuator can operate at frequency of about 0.15 Hz. The cycle duration for existing electrochemical actuators is several minutes [1, 6, 10], which corresponds to a frequency of 0.01 Hz. Thus, a short-time electrolysis produced by the alternating polarity voltage pulses demonstrates the increase of the operating frequency by more than one order of magnitude. It is worth noting that during the first three series of pulses the membrane does not reach the initial position. It returns to the initial state more slowly than expected. From the previous experiments with the fast electrochemical micropump it is known that the gas recombination time must be shorter than 1 s [9]. Slow return of the membrane can be partly connected with the formation of the pinned bubbles in the chamber, which dissolve slowly. The phenomena described above need further investigation.

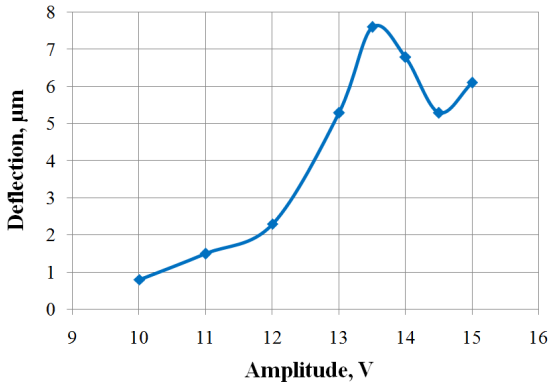

Figure 8. Dependence of the maximal membrane deflection on the amplitude of driving pulses.

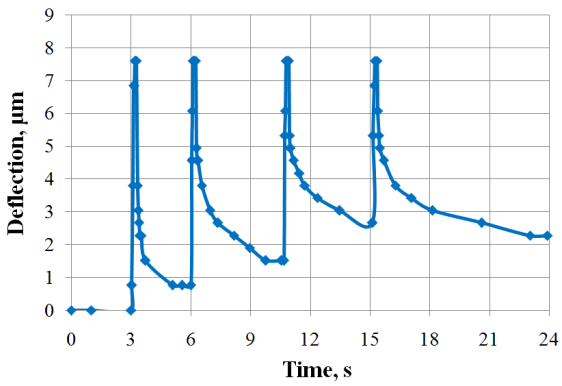

Figure 9. Deflection of the membrane with time at $U = 13.5$ V. Peaks correspond to the application of driving voltage.

Gradual wear of the electrodes is observed during the test. The working chamber after several hundred cycles is shown in figure 10. The fingers are strongly damaged. To all appearance the damage is caused by a significant energy release in the close proximity to the electrode surface as the result of the combustion reaction between hydrogen and oxygen in nanobubbles [11]. Damage is accompanied by the decrease in the Faraday current and the decrease in the membrane deflection. Such a strong wear of Ti/Al electrodes was not observed previously [9]. Probably the electrodes become less stable due to the application of SU-8 and subsequent chemical treatment. We plan to try circular electrodes similar to those used in [12]. It is expected that they will be more stable since they have less edges.

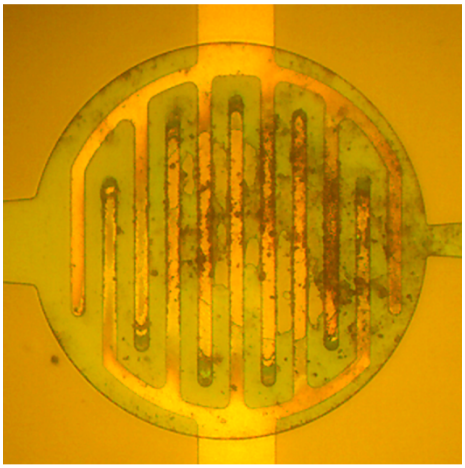

Figure 10. Working chamber after several hundred cycles.

5. Conclusions

An electrochemical membrane actuator was presented. It was designed to demonstrate a new actuation principle based on water electrolysis with a fast change of voltage polarity. The device consisted of the oxidized silicon substrate with metallic electrodes, working chamber and channels for liquid and the PDMS structure supporting the membrane. Preliminary tests of the actuator were performed, deflection of the membrane was detected using a homemade interferometer. Recently proposed alternating polarity mode allowed to achieve sufficiently short actuation cycle due to very short gas termination time in comparison with conventional electrochemical actuators. However, the cycle was longer than expected. A probable reason is the formation of pinned bubbles in the chamber, which dissolve for a relatively long time. The other problems are fast degradation and oxidation of the electrodes. An additional work has to be done to choose an appropriate material and shape for the electrodes.

Acknowledgments

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